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Post-growth thermal treatment of self-assembled InAs/GaAs quantum dots

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Abstract

Results of a post-growth rapid thermal annealing (RTA) on GaAs proximity-capped structures with high density ($\sim 10^{11}$ cm^{-2}) of self-assembled InAs/GaAs quantum dots (QDs) are presented. Features due to the QDs, bi-dimensional platelets (2DP) and InAs wetting layer (WL) were identified in photoluminescence (PL) spectrum of the as-grown sample. It is shown, using transmission electron microscopy, that RTA at temperature up to 700 °C (for 30 s) results in an increase of QDs lateral sizes. After RTA at 800 °C or higher temperatures, no QDs can be distinguished and substantial thickening of the WL can be seen. The main PL peak blueshifts as a result of RTA in all investigated temperature ranges, which is accompanied by a quenching of the 2DP and WL PL. It is proposed that the main PL peak, which is due to the QDs in the as-grown sample, results from optical recombination in the modified WL in the samples, after RTA at 800 °C and higher temperatures. Laterally-enhanced Ga/In interdiffusion induced by strain is proposed to explain a relatively fast dissolution of QDs. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Self-assembled quantum dots; Thermal treatment; Intermixing

1. Introduction

One of the ways of tuning the properties of semiconductor quantum dots (QDs) [1,2] is their postgrowth thermal treatment [3–15]. The postgrowth annealing is usually performed in a growth chamber [4,8,15] or in a rapid thermal annealing (RTA) system with SiO_2 capping [5,9,12–14] or proximity capping, (which means an annealing with a piece of GaAs substrate in close proximity to the sample surface) [3,6,7]. It is also known that RTA of samples without any capping leads to surface degradation, due to As outdiffusion [5]. Thermally-induced intermixing of the QDs leads to a blueshift, a narrowing of optical emission from QDs as well as to a reduction in the QDs interlevel spacing. It is well established that the sample capping during the thermal cycling of strained InGaAs/GaAs multi quantum wells can critically influence the results of the annealing procedure [16], however, comparative studies of the effect on thermal annealing of QDs have been

published just recently [12,13]. In this paper, the results of the effect of RTA on InAs/GaAs QDs with GaAs proximity capping are presented and discussed [17].

2. Experimental procedure

The structure investigated was grown by metaloorganic vapor phase epitaxy in the Institute of Experimental Physics, Warsaw University. The GaAs buffer layer was grown at 650 °C on a nominaly (100) oriented GaAs substrate. Self-assembled InAs QDs were formed in Stranski-Krastanow mode by growing nominally 2ML of InAs at temperature 470 °C. Then, the temperature was raised to 500 °C, while growing 100 nm of the GaAs capping layer. Samples cut from the wafer were proximity capped and RTA treated in nitrogen ambient for 30 s at 600, 700, 800, 850, 900 and 950 °C. A reference sample with wetting layer (WL) and no QDs was also grown, and RTA treated under proximity cap for 30 s at 700, 750, 800, 850 and 900 °C. The surfaces of investigated samples were not affected during the RTA procedure. The microstructures of as-grown and all annealed samples with QDs were studied using transmission electron microscopy (TEM). Cross-section-

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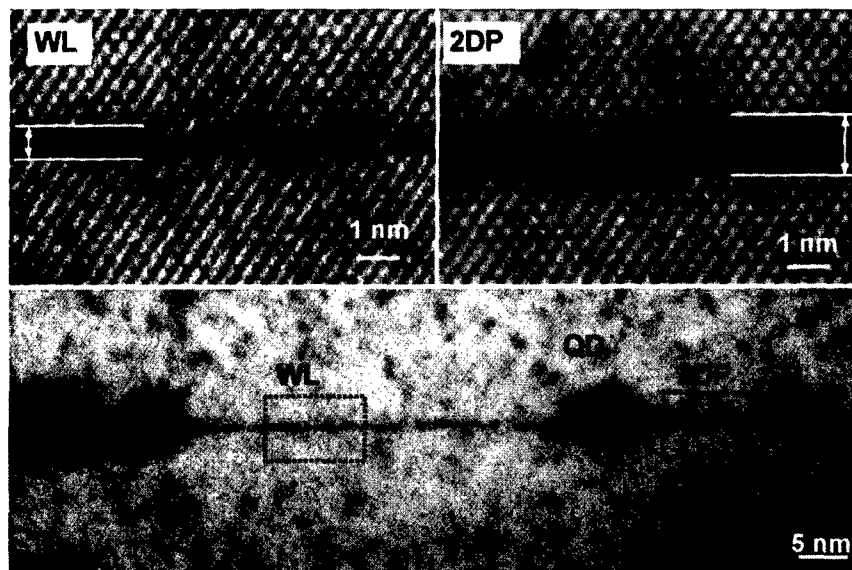E-mail address: babinski@polycnrs-gre.fr (A. Babinski).

Fig. 1. High resolution cross-sectional electron microscopy images of the as-grown sample: InAs quantum dots (QDs), InAs wetting layer (WL) and InAs bi-dimensional platelets (2DP) are identified.

al TEM specimens were prepared from the samples by a standard method of mechanical thinning, followed by ion milling. In addition, plan-view specimens were prepared from the as-grown sample. Photoluminescence (PL) measurements of both the as-grown and all annealed samples were performed at liquid-helium temperature, with laser excitation ($\lambda = 780$ nm). The signal was dispersed by a monochromator and collected onto a liquid-nitrogen-cooled germanium detector. Standard lock-in technique was applied.

3. Results

The presence of high-density self-assembled QDs in the as-grown sample was confirmed by plan-view TEM. The average size of QDs was approximately 10 nm. Their density, as estimated from the plan-view observation, was of the order of 10^{11} cm^{-2}. There was no indication of any strain relaxation in the form of large, dislocated islands or dislocations threading through the capping layer.

The QDs can be seen in the cross-sectional TEM image of the as-grown sample (Fig. 1). A 1-ml-thick InAs wetting layer (WL) can also be seen in Fig. 1. Moreover, a presence of bi-dimensional platelets (2DP) can be detected in Fig. 1. The 2DP are 2-ML-thick and they form during the growth of QDs, just before the 2D–3D transition. It has been proposed that transport and accumulation of atoms from a few 2DP leads to formation of one QD [18]. Similar coexistence of planar and three-dimensional QDs has also been reported in CdSe/ZnSe QDs system [19]. The morphology of the as-grown sample was confirmed by PL measurements. The PL spectrum (Fig. 2) of the as-grown sample is

dominated by two peaks, which are due to optical recombination within the InAs QDs ($E = 1.155$ eV) and within the InAs WL ($E = 1.42$ eV) [20]. A PL feature at $E = 1.34$ eV can also be seen in Fig. 2. Its attribution to optical recombination within the 2DP is confirmed by a band-filling effect observed in Fig. 2. No band-filling of the QDs was observed, which was probably due to their high surface density.

The RTA results in substantial changes of the sample microstructure (Fig. 3). Well-defined QDs can still be seen in samples annealed at 600 and 700 °C. The lateral sizes of the QDs increase with increasing temperature
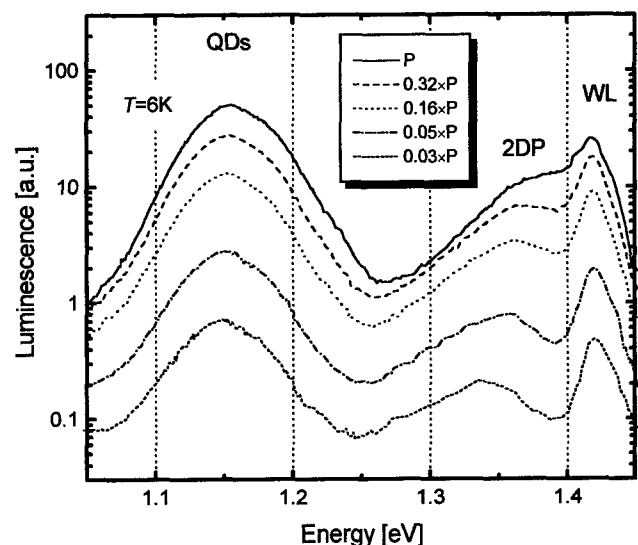

Fig. 2. Power dependence of the photoluminescence ($T = 4.2$ K) from the as-grown InAs/GaAs QDs. Note the band-filling effect on the 2DP-related PL feature [20].
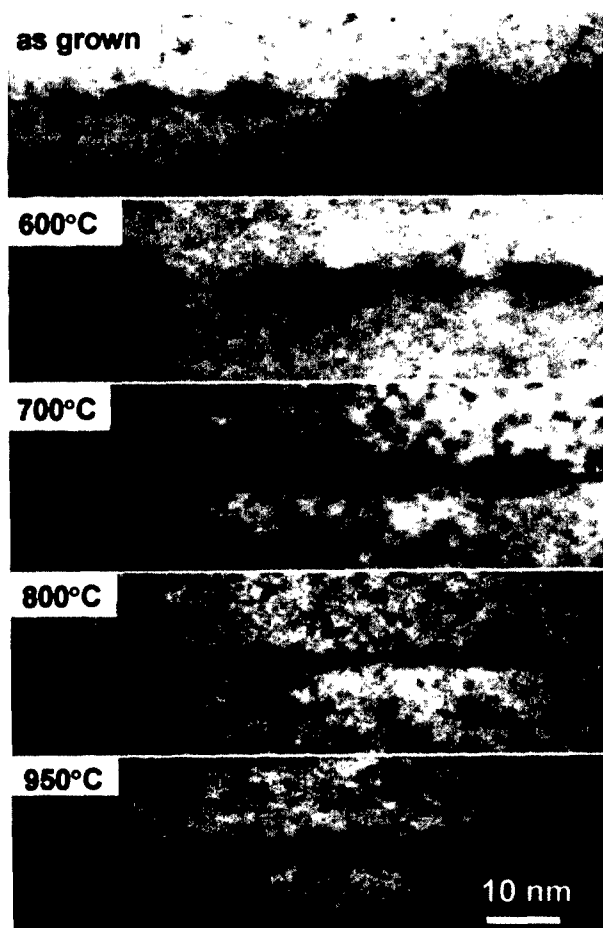
RTA under proximity capping

Fig. 3. High resolution cross-sectional TEM images of the as-grown sample and samples annealed for 30 s at 600, 700, 800 and 950 °C.

in those samples. In samples annealed at 800 °C or higher temperatures, no QDs were observed. A broadening of the modified WL was observed instead. The layer thickness increases to approximately 3 nm after annealing at 800 °C and to approximately 8 nm after annealing at 950 °C.

RTA-induced changes in the microstructure can be compared with the results of the PL measurements (Fig. 4). Annealing at 600 °C (for 30 s) results in no significant change to the QDs PL peak position (with a slight decrease of its intensity) and in a substantial decrease of the WL and 2DP PL intensity. Further changes to the PL spectrum can be observed after thermal treatment at 700 °C or higher temperatures. The RTA treatment of the samples results in a quenching of the 2DP and WL PL. Spectral position of a weak feature due to optical recombination in the WL seen after RTA at 700 °C confirms an existence of the WL of the same thickness and composition as in the as-grown sample. One peak dominates the PL spectra for samples annealed

at $T \geq 700$ °C, with the peak energy shifting from 1.33 eV (700 °C) to 1.427 eV (950 °C). In addition, a significant narrowing and intensity increase of this PL peak takes place with the increase of the annealing temperature.

A weak shift in the optical emission energy as a result of the RTA was observed in the PL spectrum of the WL sample (Fig. 5). This was accompanied by a decrease in the WL PL intensity after the RTA at highest temperatures.

4. Discussion

As it can be seen from the TEM pictures, the RTA at lowest temperatures (600 and 700 °C) results in the intermixing of the QDs. Mainly, the lateral sizes of the QDs increase as a result of that process. This must be accompanied by a change in the QDs composition, which results in a shallower potential and the PL blueshift. Simultaneously, an existence of a thin WL PL is confirmed by both TEM and PL results. After RTA at 800 °C, the QDs cannot be distinguished in the TEM images and both 2DP and WL PL peaks quench. This suggests that the QDs dissolve into the surrounding WL. The main PL peak seen from that sample must be due to the modified WL. This layer, enriched with indium from the QDs, broadens with increasing RTA temperature. Diffusion coefficients for the intermixed WLs,
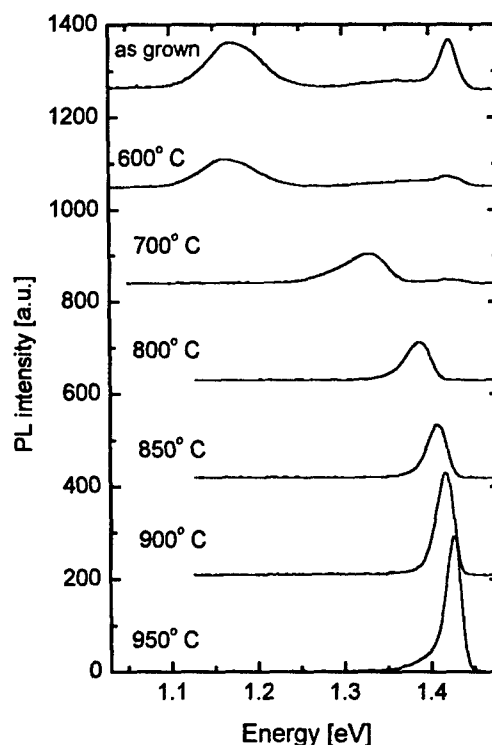

Fig. 4. Photoluminescence ($T=4.2$ K) from the as-grown and annealed samples with InAs/GaAs QDs.
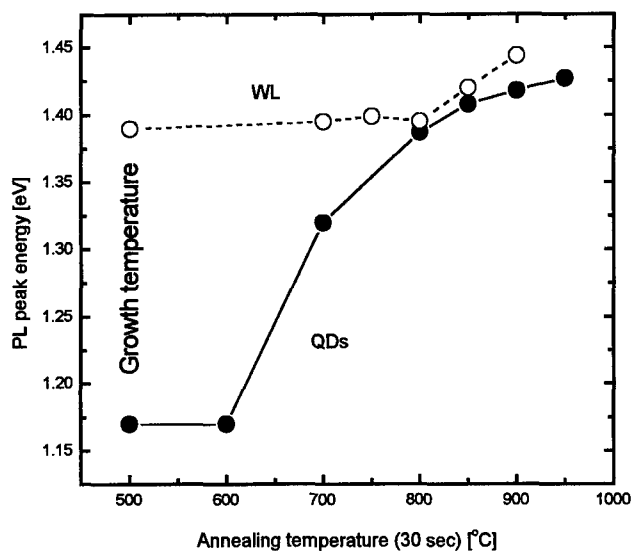
Fig. 5. Photoluminescence peak energy position as a function of annealing temperature in the samples with the InAs/GaAs QDs and the InAs WL.

estimated from their thickness measured by TEM $(2.3 \times 10^{-16}$ cm^2/s, at 800 °C to 4.1×10^{-16} cm^2/s at 900 °C and 1.7×10^{-15} cm^2/s at 950 °C) were similar to those reported for GaAs proximity-capped intermixed In$_{0.35}$Ga$_{0.65}$As/GaAs multi-quantum wells [16].

A dissolution of QDs after the RTA at high temperature (850 °C for 50 s [9] or 950 °C for 30 s [5]) has previously been observed in SiO$_2$-capped samples with InAs/GaAs QDs. That was, however, accompanied by a substantial degradation in material quality, which has not been observed in our samples. On the other hand, it has been shown that the three-dimensional character of the QDs confinement potential could be retained even after RTA at a relatively high temperature (30 s at 900 °C [5] or at 950 °C [14] and 60 s [13] at 900 °C). In our opinion, two factors are responsible for the relatively fast dissolution of the QDs in the investigated samples. These are: the GaAs proximity capping during the RTA procedure and high density of QDs.

It is well known that the interdiffusion process strongly depends on the density of such defects as vacancies, interstitials and dislocations. A number of Ga vacancies available for In/Ga interdiffusion in proximity-capped samples are limited to those existing in the sample. This can explain a weaker blue-shift of the QDs PL as a result of (nominally) defect-free interdiffusion in SrF$_2$-capped samples, as compared to that in SiO$_2$ defect-induced interdiffusion reported in [13]. Relatively low number of Ga vacancies in the proximity-capped structures results in a weak In/Ga interdiffusion [16] across the dot/cap interface, which is known to be a main mechanism of the RTA-induced PL blueshift in SiO$_2$-capped structures with QDs. Another process can be of

importance during RTA treatment of proximity-capped structures with QDs. This is the non-Fickian strain-enhanced interdiffusion [21], which has already been suggested as an explanation of some results of RTA treatment of InGaAs/GaAs QDs [3,7]. There is a large strain in and around QDs [22]. Moreover, it has recently been shown that strain fields due to lattice mismatch extend from the QDs into surrounding GaAs matrix over ≥ 10 nm [23]. Such a strain field increases vacancy concentration [23] and is likely to enhance the lateral In/Ga interdiffusion. In such a process, In atoms would replace Ga atoms in highly strained region around the QDs base. Such a behavior has already been reported in the uncapped InP/GaAs system [24]. Laterally enhanced interdiffusion could cause relatively fast dissolution of the QDs. After RTA at 600 and 700 °C, an increase in QDs lateral sizes and changes in their composition, result in a blueshift of the main PL peak. Simultaneously, sample areas covered with 1-ML-thick WL decrease, which might explain the quenching of the WL emission. Further increase in QDs lateral sizes, which occurs after RTA at higher temperatures, cancels the lateral confinement of carriers. A blueshift of the PL peak, observed after RTA in this temperature region, is due to an intermixing of the modified WL, which is enriched by indium from dissolved QDs. This blueshift is similar to that of the PL peak in the RTA samples with WL only (see Fig. 5). This must result from low Ga/In interdiffusion coefficients, characteristic for proximity-capped samples.

On the contrary, in the SiO$_2$-capped samples, a large number of Ga vacancies form at the SiO$_2$/GaAs interface during RTA. This is due to a fast Ga outdiffusion into the SiO$_2$ layer during thermal annealing [25]. Those Ga vacancies diffuse into the QDs region during RTA, enhancing the In/Ga interdiffusion through a dot/cap interface [12,13]. That process mainly affects the QDs height, which results in a blue-shift and narrowing of an optical emission, with no destruction of the three-dimensional carrier confinement in the QDs. It is very likely that the density of Ga vacancies introduced during RTA with SiO$_2$ capping is much larger than the concentration of strain-induced vacancies around the QDs (especially in the case of low-density QDs). This would explain presumably weaker lateral intermixing and preserve the three-dimensional confinement in SiO$_2$-capped RTA QDs reported in the literature. It has to be noted that some previous reports confirm the three-dimensional confinement in InGaAs/GaAs QDs after RTA under proximity capping [6]. In our opinion, this points out the role of stress in the intermixing process. InGaAs/GaAs QDs investigated in the cited work were larger (30 nm) and their surface density was lower (4×10^8 cm^{-2}) than in the case of InAs QDs investigated in our report. Both factors are likely to decrease a strain in the

structures, which might explain the difference in the annealing results.

5. Conclusions

In conclusion, we studied the effect of thermal treatment on the proximity-capped structures of self-assembled InAs/GaAs QDs and InAs WL. TEM shows a complete dissolution of QDs into surrounding WL after RTA at $T = 800$ °C for 30 s. Substantial thickening of the modified WL is observed after RTA at higher temperatures. It has been shown that the blueshift of the PL peak accompanies those morphology changes. It is proposed that the strain-enhanced lateral In/Ga interdiffusion is responsible for the fast disappearance of QDs in the investigated samples. This process may be of importance in proximity-capped annealing, when no group III vacancy formation takes place at the sample/capping interface.

Acknowledgments

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